

Regimes of correlated hopping via a two-site interacting chain

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Inelastic transport of electrons through a two-impurity chain is studied theoretically with account of intersite Coulomb interaction, U . Both limits of ohmic transport (at low bias) and strongly non-ohmic transport (at high bias) are considered. We demonstrate that correlations, induced by a finite U , in conjunction with conventional Hubbard correlations, give rise to a distinct transport regime, with current governed by *two-electron* hops. This regime realizes when a single-electron hop onto the chain and a single-electron hop out of the chain are both “blocked” due to the finite U , so that conventional correlated sequential transport is impossible. The regime of two-electron hops manifests itself in the form of an additional step in the current-voltage characteristics, $I(V)$.

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I. INTRODUCTION

A model of electron transport via two localized states positioned in sequence has been studied theoretically for more than two decades^{1–8}. There are several reasons why this model attracts the attention of researchers: (*i*) the model is tractable analytically; (*ii*) it yields nontrivial predictions that are in agreement with experimental observations^{9–15}; (*iii*) it captures physics that is much richer than tunnel transport through a single site. In particular, the two-site model allows one to reveal various aspects of *correlated* transport, that are relevant for the bulk systems, and to treat correlated transport *exactly*.

In general, for the bulk hopping conductivity, two mechanisms are known to cause correlation in the time moments of different hops, thus giving rise to the correlated character of transport.

The first mechanism is the *on-site* Hubbard repulsion, U_0 . In the limit $U_0 \rightarrow \infty$, the occupancy of a given site is restricted to the values of 0 or 1. The resulting correlation (dubbed as Hubbard correlation¹⁶) reduces to the requirement that electron hops occur only between singly occupied and empty sites.

Calculation of the current through a two-site chain with Hubbard correlations was carried out in Refs. 1, 4, 5, 7 both for ohmic^{1,4} and strongly non-ohmic^{5,7} regimes. These calculations, being exact, allow one to test the applicability of a standard mean-field description¹⁶ of the hopping transport. Within this description, the time moments of different hops are assumed completely uncorrelated, while the probability of a given hop is determined by *average* occupations of the constituting sites.

For finite U_0 , double occupation of a single site becomes possible. Together with suppression of the Hubbard correlations, finite U_0 makes the transport through a chain spin-dependent^{6,11}. This is because the second electron hopping on a given singly occupied site must, by virtue of the Pauli principle, have the opposite spin.

The second mechanism, that causes correlation be-

tween different hops is the *intersite* Coulomb interaction, U . Due to a finite U , the energy position of a given site, and, thus, the probability of an electron hop onto this site, depends on the occupation of the neighboring sites. The energy U can be viewed as an additional *charging energy* required for an electron to hop onto and out of the two-site chain.

On average, such a charging, being analogous to the Coulomb blockade, impedes the hopping transport. For bulk systems this mechanism is commonly accounted for within the mean-field theory¹⁶. The key assumption adopted to incorporate charging into the standard scheme of calculation of the hopping conductivity is that this charging amounts exclusively to the depletion of the density of states near the Fermi level¹⁷. Under this assumption, strongly correlated time evolution of the populations of *all* sites is replaced by completely uncorrelated evolution of populations of *much fewer* sites, the argument used for such a replacement being that only the hops between these few sites govern the transport.

Another effect of the intersite Coulomb interaction, which had received much less attention, is that this interaction opens the possibility for *many-electron* hops¹⁸. In the course of such hops, two (or more) electrons change their spatial positions upon absorption (or emission) of a *single* phonon. This process is analogous to the light absorption in the helium atom¹⁹, in course of which two electrons can be excited by one photon. Obviously, such an absorption is possible only due to the electron-electron interactions. It is also apparent that, as a new transport channel, many-electron hops *facilitate* the transport.

In the early paper Ref. 18, where many-electron hops were first treated analytically, the roles of the two electrons, participating in the two-electron transition, were very dissimilar. While the transfer of the first electron occurred between the sites belonging to the current-carrying network, the initial and final states of the second electron did not belong to the network. Consequently, facilitation of transport by two-electron transitions amounted to the effective reduction of the activation energies of single-electron hops within the current-

carrying network.

Later, on the basis of numerical simulations^{20,21} it was concluded that many-electron hops may constitute a significant portion of the current-carrying path through the sample. However, no analytical theory of hopping transport with account of many-electron transitions has been developed so far. This is because all existing theories are based on introducing effective resistors between the pairs of sites, which is impossible in the presence of many-electron transitions.

In the present paper, we take advantage of the fact that the two-site model is exactly solvable and thus allows one to study the interplay of all three correlation effects, namely, Hubbard correlations, Coulomb-blockade-induced correlations, and correlated two-electron hops. The fact that the first two correlation mechanisms impede the transport, while the third one facilitates it, suggests that such an interplay is nontrivial.

Our main result is a demonstration of a distinct regime of transport through the two-site chain, in which two-electron hops *dominate* the passage of current. This regime becomes possible due to intersite interaction, U , when both manifestations of this interaction are at work: (*i*) finite U allows two-electron transitions, and (*ii*) it blocks both individual single-electron transitions onto and out of the chain. Conceptually, this interaction-dominated transport regime is analogous to inelastic cotunneling through a Coulomb blockaded quantum dot^{22–24}. However, in terms of its manifestations, the important difference between the dot and the two-site chain is that the dot contains many levels, so that the cotunneling rate is a sum of many contributions. By contrast, the transport through a blocked two-site chain is governed by a single two-electron hop, leading to distinctive temperature and bias dependencies of current.

We first identify the regime of two-electron hops for strongly non-ohmic transport at zero temperature, where it manifests itself in the form of additional steps in the current-voltage characteristics, $I(V)$. Then we demonstrate the relevance of two-electron hops for the ohmic transport.

II. HOPPING TIMES

The two-site model is illustrated in Fig. 1. Under the applied bias, V , the Fermi levels in the left and right leads are shifted $V/2$ and $-V/2$, respectively. The two sites, 1 and 2, are located at distances d_1 and d_2 with respect to the center of the barrier of thickness, D . We adopt the definition of the energy position of site 1 to be ε_1 , when site 2 is *empty*. Consequently, when site 2 is occupied, the energy position of site 1 is $\varepsilon_1 + U$. Analogously, ε_2 and $\varepsilon_2 + U$ are the energy positions of site 2 for empty and occupied site 1, respectively. We assume that there is tunnel coupling with amplitude, $t_{1,k}$, between site 1 and the extended state, \mathbf{k} , in the left lead.

Similarly, the state, \mathbf{p} , in the right lead is coupled to site 2 with amplitude, $t_{2,p}$. Then the waiting time, τ_1 , for an electron in the left lead to tunnel onto site 1 is given by $\tau_1^{-1} = f_l(\varepsilon_1)\Gamma_l(\varepsilon_1)/\hbar$, where the tunneling width, $\Gamma_l(\varepsilon)$, is defined as

$$\Gamma_l(\varepsilon) = 2\pi \sum_{\mathbf{k}} |t_{1,k}|^2 \delta(\varepsilon - \varepsilon_{\mathbf{k}}) \propto \exp\left[-\frac{D - 2d_1}{a}\right]. \quad (1)$$

The corresponding expression for the waiting time, τ_3 , for electron on site 2 to escape into the right lead reads $\tau_3^{-1} = [1 - f_l(\varepsilon_2)]\Gamma_r(\varepsilon_2)/\hbar$, with

$$\Gamma_r(\varepsilon) = 2\pi \sum_{\mathbf{p}} |t_{2,p}|^2 \delta(\varepsilon - \varepsilon_{\mathbf{p}}) \propto \exp\left[-\frac{D - 2d_2}{a}\right]. \quad (2)$$

Here a is the localization radius of the on-site wave functions; f_l , f_r are the Fermi distribution functions in the left and right leads, respectively, which determine the temperature dependencies of τ_1 and τ_3 .

Transition $1 \rightarrow 2$ involves tunneling accompanied by the emission of a phonon with energy $\varepsilon_1 - \varepsilon_2$. Thus, the temperature dependence of the corresponding time, $\tau_2 \propto \exp[2(d_1 + d_2)/a]$, is weak.

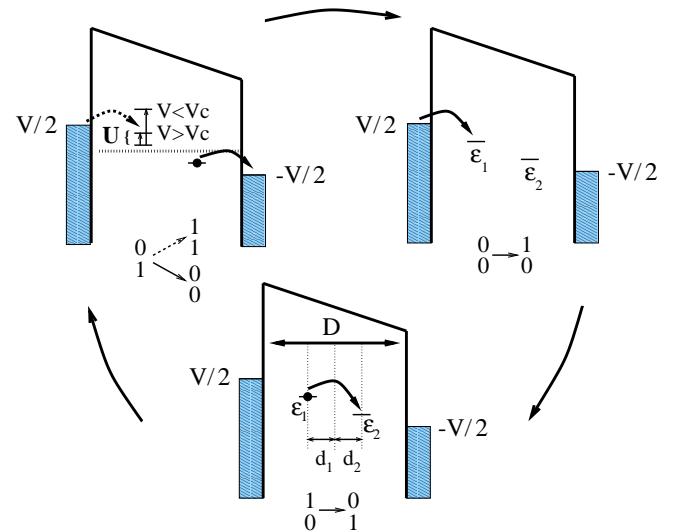


FIG. 1. Schematic illustration of the two-site model. Evolution of the occupation numbers of sites in the sequential hopping regime for blocked ($V < V_c$) and unblocked ($V > V_c$) transition $l \rightarrow 1$.

In all previous analytical treatments^{1–8} of the two-site model it was assumed that the passage of current is governed by only three transitions: $l \rightarrow 1$, $1 \rightarrow 2$, and $2 \rightarrow r$. Our key point in this study is that the tunnel couplings $t_{1,k}$ and $t_{2,p}$ together with finite intersite interaction, U , allow for the additional transition, $(l, 2) \rightarrow (1, r)$, that involves simultaneous change of occupation of *both* sites. Moreover, we will demonstrate that, within a certain bias range, these two-electron transitions dominate the passage of current through the two-site chain. Calculation

of the waiting time, τ_c , for the two-electron transition is similar to the calculation²⁴ of the inelastic cotunneling rate through a quantum dot

$$\frac{1}{\tau_c} = \frac{2\pi}{\hbar} \sum_{\mathbf{k},\mathbf{p}} |t_{1,k} t_{2,p}|^2 \left[\frac{1}{\varepsilon_2 - \varepsilon_p} + \frac{1}{\varepsilon_k - \varepsilon_1 - U} \right]^2 \times \delta(\varepsilon_k + \varepsilon_2 - \varepsilon_p - \varepsilon_1) f_l(\varepsilon_k) [1 - f_r(\varepsilon_p)], \quad (3)$$

where the two terms in the square brackets originate from the intermediate states with empty and doubly-occupied chain, respectively. The many-body nature of the two-electron transition manifests itself in the fact that these terms cancel each other in the limit $U \rightarrow 0$.

With the help of definitions (1) and (2), Eq. (3) can be presented as

$$\frac{1}{\tau_c} = U^2 \int d\varepsilon \frac{\Gamma_l(\varepsilon_1 - \varepsilon)\Gamma_r(\varepsilon_2 - \varepsilon)}{2\pi\hbar\varepsilon^2(\varepsilon + U)^2} f_l(\varepsilon_1 - \varepsilon) [1 - f_r(\varepsilon_2 - \varepsilon)]. \quad (4)$$

In principle, the integrand needs to be regularized at the resonances, $\varepsilon = 0, -U$, following, *e.g.*, the procedure of Ref. 25. However, the corresponding singular contributions are negligibly small in the cases of interest, $|\varepsilon_1|, |\varepsilon_2| \gg T$, where T is the temperature.

We will establish the analytical form of τ_c in the limit of large enough bias, V , when it is temperature-independent. In this limit, the integration in Eq. (4) is restricted to the interval $\varepsilon_1 - V/2 < \varepsilon < \varepsilon_2 + V/2$, which is set by the distribution functions in the leads. Neglecting the weak energy dependencies of Γ_l and Γ_r , and evaluating the integral, we obtain

$$\frac{1}{\tau_c} = \frac{\Gamma_l \Gamma_r}{\pi \hbar U} \left[\Phi\left(\frac{2\varepsilon_1 - V}{2U}\right) - \Phi\left(\frac{2\varepsilon_2 + V}{2U}\right) \right], \quad (5)$$

where the function $\Phi(x)$ is defined as

$$\Phi(x) = \frac{2x+1}{2x(x+1)} - \ln\left(\frac{x+1}{x}\right). \quad (6)$$

The asymptotes of τ_c in the limits of small and large intersite interaction, U , can easily be found from Eqs. (5), (6)

$$\frac{1}{\tau_c} = \frac{4\Gamma_l \Gamma_r U^2}{3\pi\hbar} \left[\frac{1}{(2\varepsilon_1 - V)^3} - \frac{1}{(2\varepsilon_2 + V)^3} \right], \quad U \rightarrow 0, \quad (7)$$

$$\frac{1}{\tau_c} = \left(\frac{2\Gamma_l \Gamma_r}{\pi\hbar} \right) \frac{\varepsilon_2 - \varepsilon_1 + V}{(2\varepsilon_1 - V)(2\varepsilon_2 + V)}, \quad U \rightarrow \infty. \quad (8)$$

It is worth noting that the rate Eq. (3) of the interaction-induced hop becomes U -independent in the limit of

strong intersite interaction, as follows from Eq. (3) as well as from Eq. (8). Note also that in both limits, the rate τ_c^{-1} is proportional to $(\varepsilon_2 - \varepsilon_1 + V)$, which plays the role of “phase volume” for two-electron transitions, as we will see below.

It is seen from Eq. (3) that $\tau_c \propto \exp[2(D - d_1 - d_2)/a]$, i.e. it is shorter than the waiting time for direct tunneling, which is $\propto \exp(2D/a)$. Important, however, is that τ_c can be comparable to τ_2 . As we will see later, transport, dominated by two-electron transitions, is most prominent when τ_c and τ_2 are of the same order.

III. NON-OHMIC REGIME

A. QUALITATIVE DISCUSSION

In this Section we assume that the temperature is zero, so that hopping transport through a chain is possible if all hops, involved in the passage of current, are *activationless*. All throughout the paper we assume the on-site Hubbard repulsion, U_0 , to be infinite. To classify different transport regimes, we first note that when the bias, V , is big enough, $V \gg U$, the intersite repulsion, U , can be neglected. The current path then consists of three hops, $l \rightarrow 1$, $1 \rightarrow 2$, and $2 \rightarrow r$, in *arbitrary* order, in the sense that the only condition for a hop to occur is that the initial state is occupied, while the final state is empty. In particular, as illustrated in Fig. 1, the waiting time for the hop $l \rightarrow 1$ does not depend on whether site 2 is occupied or empty. Similarly, the waiting time for the hop $2 \rightarrow r$ is independent of the occupation of site 1. This, however, does not mean that the transport in the regime of high bias is completely uncorrelated. It only means that the correlations are of purely Hubbard origin, i.e. the electron in the left lead has to “wait” for the hop $1 \rightarrow 2$, leaving site 1 empty, to occur. This Hubbard-correlated regime takes place for $V > V_c$, where V_c is determined by the condition $\varepsilon_1 + U = V_c/2$. As seen from Fig. 1, at $V = V_c$, site 1, shifted upward by U , due to site 2 being occupied, becomes aligned with the Fermi level in the left lead.

As the bias is reduced below V_c , the intersite repulsion changes radically the electron dynamics. Now the activationless hop $l \rightarrow 1$ is possible *only* if site 2 is empty (see Fig. 1). When site 2 is occupied, the transition $l \rightarrow 1$ is “blocked”. Clearly, the average current drops down in a step-like fashion as the bias is swept through V_c . It is also clear that below V_c the current is more correlated than above V_c . This is because the hops $l \rightarrow 1$, $1 \rightarrow 2$, and $2 \rightarrow r$ occur in a strict succession.

The two above regimes were discussed in the literature before^{5,7}. However, the most nontrivial scenario of activationless passage of current unfolds when *both* transitions $l \rightarrow 1$ and $2 \rightarrow r$ are blocked. The concept of inelastic cotunneling through a dot^{22–24} suggests that in this case the activationless current is due to the two-electron

transition $(l, 2) \rightarrow (1, r)$. However, the theory of inelastic cotunneling implies that the two-electron transition is *immediately* followed by *relaxation*. For a two-site chain this relaxation is a single-electron transition $1 \rightarrow 2$. Then the “immediate” relaxation requires that sites 1 and 2 are spatially close to each other (as is the case for dots). Note, however, that the spatial proximity of sites 1 and 2, meaning that $d_1, d_2 \ll D$, makes the two-electron time $\tau_c \propto \exp[2(D - d_1 - d_2)/a]$ {see Eq. (3)} quite long, and actually comparable to the direct-tunneling time. Thus, in order to yield a significant current, the separation, $(d_1 + d_2)$, should be $\sim D$. On the other hand, large separation of sites 1 and 2 unavoidably opens up new relaxation channels, which are “reverse” single-electron transitions $1 \rightarrow l$ and $r \rightarrow 2$. Obviously, these transitions “undo” the two-electron transition and forbid the current to flow. Then, it is quite nontrivial that for certain energy configurations $\{\varepsilon_1, \varepsilon_2\}$ of the sites, the reverse relaxation channels are blocked, leaving the transition $1 \rightarrow 2$, the *only* allowed transition that can follow a two-electron hop. At the same time, the transition $1 \rightarrow 2$, happening after the two-electron hop, *completes the current cycle*. We will dub such configurations with unidirectional relaxation as ratchet configurations.

B. RATCHET-TYPE CONFIGURATIONS

Consider the configuration of sites depicted in Fig. 2. Site 2 is lower than $-V/2$ and thus is occupied. The energy, $\varepsilon_1 + U$, of site 1 is above $V/2$, so that site 1 is empty. For this configuration both transitions $l \rightarrow 1$ and $2 \rightarrow r$ are blocked. The condition that the two-electron transition $(l, 2) \rightarrow (1, r)$ is allowed at zero temperature reads

$$\frac{V}{2} - \left(-\frac{V}{2}\right) = V > (\varepsilon_1 - \varepsilon_2). \quad (9)$$

The lhs in Eq. (9) is the minimal energy required to transfer an electron between the leads, while the rhs is the energy required to transfer an electron between sites 2 and 1.

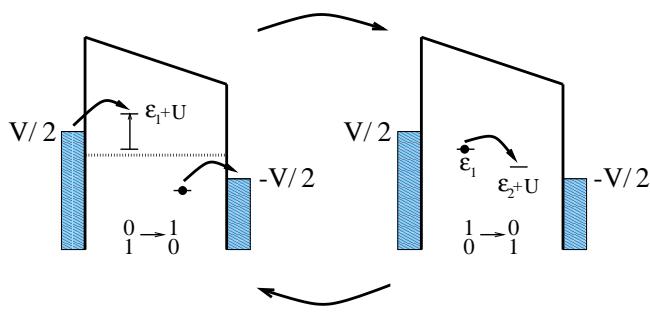


FIG. 2. Schematic illustration of the non-ohmic transport regime dominated by two-electron hops $(l, 2) \rightarrow (1, r)$ followed by an inelastic transition $1 \rightarrow 2$. For the ratchet-type configuration shown, both “backwards” transitions following $(l, 2) \rightarrow (1, r)$ are forbidden.

The condition allowing a two-electron transition has the form Eq. (9) since this transition can be viewed as a transfer of an electron from the left to the right lead, accompanied by a “backward” excitation $2 \rightarrow 1$ of the other electron. Obviously, the “direct” transition $1 \rightarrow 2$ can occur after the two-electron hop. The ratchet-type configuration is such that this direct transition is not preceded by other single-electron transitions. This would absolutely be the case if the following two conditions are met

$$\varepsilon_1 < \frac{V}{2}, \quad (10)$$

$$\varepsilon_2 + U > -\frac{V}{2}. \quad (11)$$

Indeed, the first condition ensures that the electron from the occupied site 1 cannot hop back into the left lead, while the second condition guarantees that the empty site 2 does not get occupied as a result of an electron hop from the right lead. Our prime observation is that ratchet-type configurations are possible, i.e. all five conditions, $(\varepsilon_1 + U) > V/2$, $\varepsilon_2 < -V/2$, and Eqs. (9)-(11), are satisfied simultaneously within a *finite* domain on the $(\varepsilon_1, \varepsilon_2)$ plane. This domain corresponds to the triangular region in Fig. 3, where I_c denotes the magnitude of current in the regime of two-electron hops. This region is situated adjacently to two rectangular domains, within which one of the transitions $l \rightarrow 1$ or $2 \rightarrow r$ is blocked. I_1 stands for the magnitude of the strictly sequential current in these domains. Finally, within the domain $(\varepsilon_1 < -U + V/2, \varepsilon_2 > -V/2)$ both transitions are unblocked, so that the current, I_2 , is limited only by the Hubbard correlations.

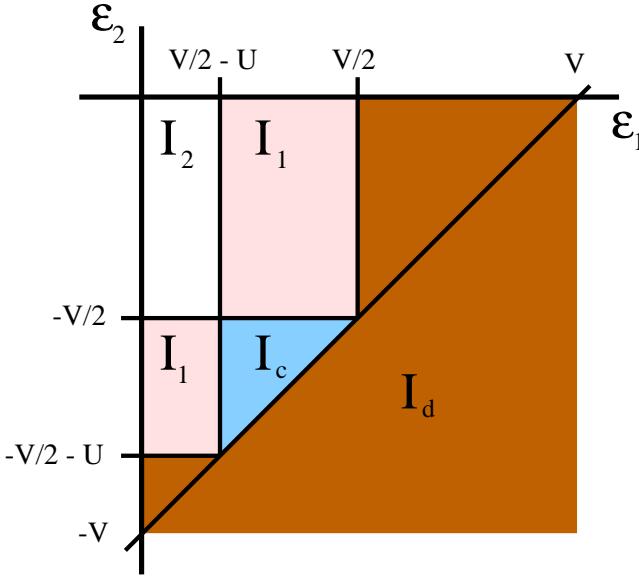


FIG. 3. (Color online) Domains of the site energies corresponding to the different regimes of transport.

To summarize our qualitative analysis, in Fig. 4 we depict schematically the current-voltage characteristics at zero temperature. In the absence of interactions the $I - V$ curve would exhibit only one step, from the “long-hop” current, I_d , corresponding to direct tunneling between the leads, to Hubbard-correlated current, I_2 . Interactions give rise to two additional steps: from I_d to “doubly-blockaded” current, I_c , and from I_c to “singly-blockaded” current, I_1 .

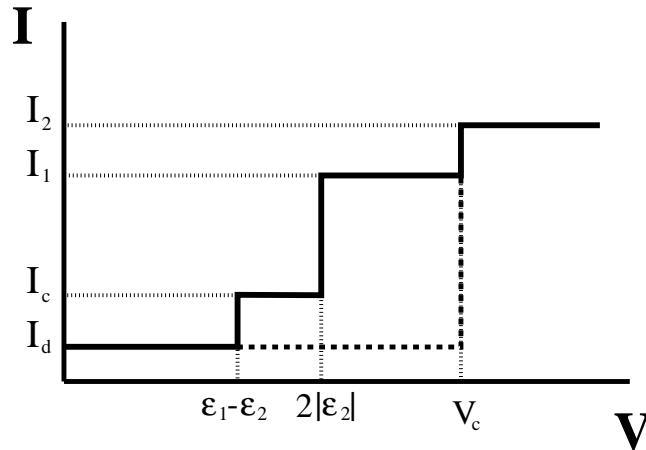


FIG. 4. Schematic $I - V$ characteristics of the two-site chain at zero temperature is shown for the case $-U < \varepsilon_1 + \varepsilon_2 < 0$. Weak bias dependencies of current within each step are neglected.

In the remainder of this Section we calculate the values I_1 , I_2 , and I_c , and study the correlation characteristics of transport within each step.

C. CALCULATION OF CURRENT

For infinite Hubbard repulsion, possible sets of the occupation numbers (n_1, n_2) of the sites 1 and 2 are restricted to $(0, 0)$, $(0, 1)$, $(1, 0)$, and $(1, 1)$. Following Refs. 1, 4 we introduce the probabilities, P_{n_1, n_2} , of each set. Normalization requires that $P_{0,0} + P_{0,1} + P_{1,0} + P_{1,1} = 1$.

As discussed above, in the regime $V > V_c$, when both transitions $l \rightarrow 1$ and $2 \rightarrow r$ are unblocked, the transport involves all four sets of the occupation numbers. For $V < V_c$, when $l \rightarrow 1$ is blocked, double occupation is prohibited, i.e. $P_{1,1} = 0$. The master equations for probabilities P_{n_1, n_2} can be cast in a form that accounts for both cases, i.e. applies within the entire interval $-V/2 < \varepsilon_2 < \varepsilon_1 < V/2$, as follows

$$\frac{dP_{1,0}}{dt} = -\frac{P_{1,0}}{\tau_2} + \frac{P_{0,0}}{\tau_1} + \frac{P_{1,1}}{\tau_3}\Theta(V - V_c), \quad (12)$$

$$\frac{dP_{0,1}}{dt} = -\frac{P_{0,1}}{\tau_3} - \frac{P_{0,1}}{\tau_1}\Theta(V - V_c) + \frac{P_{1,0}}{\tau_2}, \quad (13)$$

$$\frac{dP_{0,0}}{dt} = -\frac{P_{0,0}}{\tau_1} + \frac{P_{0,1}}{\tau_3}, \quad (14)$$

where $\Theta(x)$ is the step-function. It ensures that $P_{1,1}$ drops out of the system (12)-(14) for $V < V_c$. The expression for current that accounts for both cases reads

$$I = \frac{eP_{0,1}}{\tau_3} + \frac{eP_{1,1}}{\tau_3}\Theta(V - V_c). \quad (15)$$

In the stationary regime, the master equations can be solved for P_{n_1, n_2} in terms of $P_{0,1}$, namely, $P_{0,0} = P_{0,1}\tau_1/\tau_3$, $P_{1,0} = P_{0,1}[\tau_2/\tau_3 + (\tau_2/\tau_1)\Theta(V - V_c)]$, $P_{1,1} = P_{0,1}(\tau_3/\tau_1)\Theta(V - V_c)$. With the normalization condition, $P_{0,0} + P_{0,1} + P_{1,0} + P_{1,1}\Theta(V - V_c) = 1$, we get

$$P_{0,1} = \frac{\tau_3}{\tau_1 + \tau_2 + \tau_3 + \frac{\tau_3}{\tau_1}(\tau_2 + \tau_3)} \quad (16)$$

for $V > V_c$ and

$$P_{0,1} = \frac{\tau_3}{\tau_1 + \tau_2 + \tau_3} \quad (17)$$

for $V < V_c$. Substituting Eqs. (16), (17) into Eq. (15) yields^{5,7}

$$I_2 = \frac{e(\tau_1 + \tau_3)}{(\tau_1 + \tau_3)^2 - \tau_1\tau_3 + \tau_2(\tau_1 + \tau_3)}, \quad V > V_c, \quad (18)$$

and

$$I_1 = \frac{e}{\tau_1 + \tau_2 + \tau_3}, \quad V < V_c. \quad (19)$$

To find I_c , we note that in the regime of current dominated by two-electron transitions only $P_{0,1}$ and $P_{1,0}$ are nonzero. They are related via the master equation

$$\frac{dP_{1,0}}{dt} = -\frac{P_{1,0}}{\tau_2} + \frac{P_{0,1}}{\tau_c} \quad (20)$$

and the normalization condition $P_{0,1} + P_{1,0} = 1$. In the stationary regime, these two relations yield for $I_c = P_{1,0}/\tau_2$ the following expression

$$I_c = \frac{e}{\tau_2 + \tau_c}. \quad (21)$$

The $I - V$ characteristics of a chain contains three steps, see Fig. 4. Their magnitudes, I_c/I_d , I_1/I_c , and I_2/I_1 , depend on the spatial positions of the sites. From Eqs. (18)-(21) it is easy to analyze the dependence of these magnitudes on d_1 and d_2 . First, we note that the product $\tau_1\tau_2\tau_3 \propto \exp(2D/a)$ does not depend on d_1, d_2 .

$$\frac{I_2}{I_1} = \frac{2 \cosh\left(\frac{d_1-d_2}{a}\right) \left[2 \cosh\left(\frac{d_1-d_2}{a}\right) + \exp\left\{\frac{3}{a}\left[(d_1+d_2) - \frac{D}{3}\right]\right\} \right]}{\left[2 \cosh\left(\frac{d_1-d_2}{a}\right) \right]^2 + 2 \cosh\left(\frac{d_1-d_2}{a}\right) \exp\left\{\frac{3}{a}\left[(d_1+d_2) - \frac{D}{3}\right]\right\} - 1}. \quad (24)$$

From Eqs. (22)-(24) it is easy to see that the magnitude of the first step is large, as long as $d_1, d_2 \gg a$. Concerning the second and the third steps, the more pronounced they are, the smaller the difference $|d_1 - d_2|$. Even for a completely symmetric arrangement, $d_1 = d_2 = d$, the second step is present only if $d < D/4$. Precisely at $d = D/4$, we have $\tau_2 = \tau_c$. As d decreases, the magnitude of the second step grows first as $\exp\left\{8(D/4 - d)/a\right\}$ for $D/4 > d > D/6$, and then slower, as $\exp\left\{2(D/2 - d)/a\right\}$, for $d < D/6$. The origin of this growth is that for smaller d the waiting time, τ_c , for two-electron transitions becomes progressively longer than τ_1, τ_3 . It is also possible to conclude from Eq. (24) that the magnitude of the third step, I_2/I_1 , does not exceed 4/3. The maximum magnitude corresponds to $d_1 = d_2 < D/6$.

D. CORRELATION PROPERTIES

As mentioned in the Introduction, the advantage of the two-site model being exactly solvable is that it allows one to analyze the applicability of the mean-field approach, one of the main ingredients of the theory of hopping transport. The exact solution captures correlations in the occupation numbers of sites, whereas within the mean-field description these correlations are neglected. For the ohmic transport, the effect of correlations on the average current was studied in Ref. 1. Below we examine the correlation properties of current for various regimes of non-ohmic transport.

The average occupation numbers of the sites, $\langle n_1 \rangle$ and $\langle n_2 \rangle$, can be expressed in terms of P_{n_1, n_2} as $\langle n_1 \rangle =$

Secondly, the product $\tau_1\tau_3$ contains the same tunneling exponent as τ_c . These observations allow one to express the magnitudes of the steps as follows

$$\frac{I_c}{I_d} = \frac{\exp\left(\frac{D}{a}\right)}{2 \cosh\left\{\frac{2}{a}\left[\frac{D}{2} - (d_1 + d_2)\right]\right\}}, \quad (22)$$

$$\frac{I_1}{I_c} = \frac{1 + \exp\left\{\frac{4}{a}\left[\frac{D}{2} - (d_1 + d_2)\right]\right\}}{1 + 2 \cosh\left(\frac{d_1-d_2}{a}\right) \exp\left\{\frac{3}{a}\left[\frac{D}{3} - (d_1 + d_2)\right]\right\}}, \quad (23)$$

(24)

$P_{1,0} + P_{1,1}$ and $\langle n_2 \rangle = P_{0,1} + P_{1,1}$, respectively. Neglecting correlations is equivalent to setting zero the difference

$$\begin{aligned} \langle n_1 n_2 \rangle - \langle n_1 \rangle \langle n_2 \rangle &= P_{11} - (P_{1,0} + P_{1,1})(P_{0,1} + P_{1,1}) \\ &= P_{0,0} P_{1,1} - P_{0,1} P_{1,0}. \end{aligned} \quad (25)$$

We will characterize correlations by the parameter $\kappa = [\langle n_1 n_2 \rangle / \langle n_1 \rangle \langle n_2 \rangle] - 1$, which, using Eq. (25), can be presented as

$$\kappa = \frac{P_{0,0} P_{1,1} - P_{0,1} P_{1,0}}{\langle n_1 \rangle \langle n_2 \rangle}, \quad (26)$$

so that for uncorrelated transport $\kappa = 0$. It can now be shown that, for $V > V_c$, the *exact* equations (12)-(14) can be cast in the following form

$$\begin{aligned} \frac{d \langle n_1 \rangle}{d t} &= \frac{(1 - \langle n_1 \rangle)}{\tau_1} - \frac{\langle n_1 \rangle (1 - \langle n_2 \rangle)}{\tau_2} + \kappa \frac{\langle n_1 \rangle \langle n_2 \rangle}{\tau_2}, \\ \frac{d \langle n_2 \rangle}{d t} &= \frac{\langle n_1 \rangle (1 - \langle n_2 \rangle)}{\tau_2} - \frac{\langle n_2 \rangle}{\tau_3} - \kappa \frac{\langle n_1 \rangle \langle n_2 \rangle}{\tau_2}. \end{aligned} \quad (27)$$

In this form, the mean-field description emerges upon neglecting the last terms in the right-hand sides. Solving the resulting system of equations for average occupations, we reproduce the result of Ref. 3

$$I_2^{\text{MF}} = \frac{e \langle n_2 \rangle}{\tau_3} = \frac{2e}{\tau_1 + \tau_2 + \tau_3 + \left[(\tau_1 + \tau_2 + \tau_3)^2 - 4\tau_1\tau_3 \right]^{1/2}}. \quad (28)$$

From Eqs. (18) and (28) it can be seen that the ratio I_2/I_2^{MF} can be expressed as a function of a dimensionless parameter $z = \tau_2 (\tau_1^{-1} + \tau_3^{-1})$ as follows

$$F(z) = \frac{I_2}{I_2^{\text{MF}}} = \frac{b + z + \sqrt{(b+z)^2 - 4b}}{2(b+z-1)}, \quad b = \frac{(\tau_1 + \tau_3)^2}{\tau_1 \tau_3}. \quad (29)$$

The function $F(z)$ is plotted in Fig. 5 for different values of the ratio τ_1/τ_3 . We see that, unlike the ohmic case¹, the exact current can exceed the mean-field value if τ_2 is sufficiently long. Moreover, for $z = 1$, we have $I_2 = I_2^{\text{MF}}$ for arbitrary ratio τ_1/τ_3 .

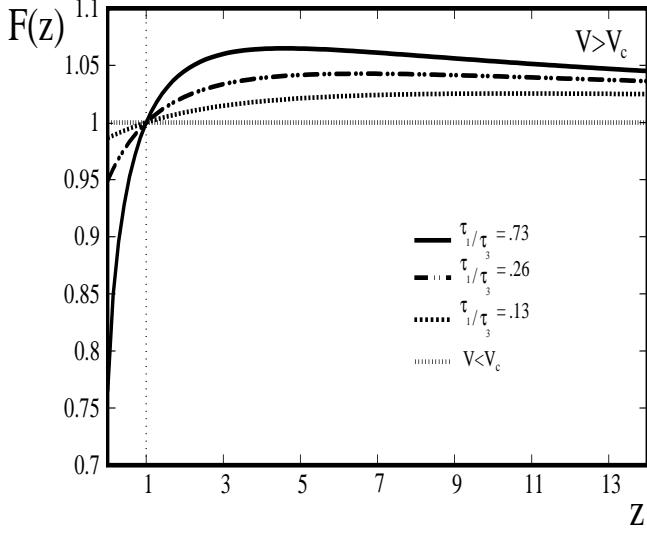


FIG. 5. Ratio of exact and mean-field currents is plotted versus a dimensionless parameter $z = \tau_2 (\tau_1^{-1} + \tau_3^{-1})$ for $\tau_1/\tau_3 = 0.73$ (solid line); $\tau_1/\tau_3 = 0.26$ (dashed-dotted line); and $\tau_1/\tau_3 = 0.13$ (dashed line).

The explanation of these facts can be obtained if we express the correlation parameter, κ , in terms of τ_1 , τ_2 , and τ_3 , using Eq. (16)

$$\kappa = \frac{\tau_1 [\tau_1 \tau_3 - \tau_2 (\tau_1 + \tau_3)]}{(\tau_1 + \tau_3)(\tau_1 \tau_2 + \tau_2 \tau_3 + \tau_3^2)} = \frac{\tau_1^2 (1 - z)}{(\tau_1 + \tau_3)(\tau_1 z + \tau_3)}. \quad (30)$$

We see that for $z = 1$ we have $\kappa = 0$, so that the current is effectively uncorrelated. For this reason we have $I_2 = I_2^{\text{MF}}$ when $z = 1$. For $z < 1$, the correlation parameter is positive, resulting in $I_2 < I_2^{\text{MF}}$. Similarly, $I_2 > I_2^{\text{MF}}$ for $z > 1$ is the manifestation of the fact that the current is *negatively* correlated. The latter has a simple explanation: for long τ_2 (i.e. for $z \gg 1$), as an electron tunnels onto site 1 from the left lead, site 2 is likely to be *empty*. Then the electron does not have to “wait” extra time beyond τ_2 to proceed to site 2. The mean-field description does not capture this effect, thus causing $I_2^{\text{MF}} < I_2$.

For $V < V_c$ the mean-field description reduces to the following equations for the average occupation numbers

$$\frac{d \langle n_1 \rangle}{dt} = \frac{(1 - \langle n_1 \rangle)[1 - \langle n_2 \rangle]}{\tau_1}$$

$$-\frac{\langle n_1 \rangle(1 - \langle n_2 \rangle)}{\tau_2}, \\ \frac{d \langle n_2 \rangle}{dt} = \frac{\langle n_1 \rangle(1 - \langle n_2 \rangle)}{\tau_2} - \frac{\langle n_2 \rangle}{\tau_3}, \quad (31)$$

where the factor $(1 - \langle n_2 \rangle)$ in the first equation expresses the fact that $l \rightarrow 1$ is possible only when site 2 is empty. It is easy to see that these equations yield $I_1^{\text{MF}} = e/(\tau_1 + \tau_2 + \tau_3)$, i.e. the *exact* value of current in the regime $V < V_c$, see Eq. (19). The explanation why neglecting the correlations does not change the average current can be inferred from rewriting Eqs. (31) in the following form

$$\frac{\tau_1}{\tau_1 + \tau_2} \frac{d \langle n_1 \rangle}{dt} = \left[\frac{(1 - \langle n_2 \rangle)}{\tau_1 + \tau_2} - \frac{\langle n_1 \rangle}{\tau_2} \right] + \frac{\langle n_1 \rangle \langle n_2 \rangle}{\tau_2}, \\ \frac{d \langle n_2 \rangle}{dt} = \left[-\frac{\langle n_2 \rangle}{\tau_3} + \frac{\langle n_1 \rangle}{\tau_2} \right] - \frac{\langle n_1 \rangle \langle n_2 \rangle}{\tau_2}. \quad (32)$$

Note now, that if one neglects the product $\langle n_1 \rangle \langle n_2 \rangle$ in the right-hand sides, then the mean-field equations (32) reduce to the *exact* equations (12)-(14) for $V < V_c$, since for $V < V_c$ we have $\langle n_1 \rangle = P_{1,0}$ and $\langle n_2 \rangle = P_{0,1}$. Then the fact that both Eqs. (32) and (12)-(14) yield the same value of $\langle n_2 \rangle$, and thus the same current I_1 , is a consequence of the observation that, upon adding the two equations (32), the products $\langle n_1 \rangle \langle n_2 \rangle$ cancel each other.

As follows from Eq. (25), in two other regimes with $I = I_1$ and $I = I_c$ we have $\kappa = -1$. This reflects the fact that the passage of current occurs via a *single* repeating cycle in both regimes. In particular, the cycle for $I = I_c$ consists of two-electron transition $(l, 2) \rightarrow (1, r)$ followed by a single-electron transition $1 \rightarrow 2$. The corresponding waiting times obey the Poisson distributions with averages τ_c and τ_2 , respectively. This allows one to find the Fano factor of the current noise in the regime $I = I_c$ to be $(\tau_c^2 + \tau_2^2)/(\tau_c + \tau_2)^2 < 1$. Current noise in the Hubbard-correlated regime, $I = I_2$, was studied in detail in Ref. 7.

IV. OHMIC REGIME

Similarly to sequential hopping, the transport regime dominated by two-electron transitions is activationless only if applied bias, V , is high enough. Indeed, as seen from Fig. 3, the domain where the current is equal to I_c exists only for finite V . As the bias is reduced, the current assumes activational character in both regimes. Then the question arises as to whether the two-electron regime survives in the ohmic limit $V \ll T$, where T is the temperature. We will address this question in the following sequence. First we derive an expression, analogous to Eq. (21), for the ohmic resistance due to two-electron transitions. Then we will demonstrate that this expression applies to the two-site chain within a certain domain of energies, $\varepsilon_1, \varepsilon_2$, and positions d_1, d_2 of the sites.

In order to generalize the derivation of Eq. (21) to the ohmic case, we introduce, along with the time τ_2 , the time of a “reverse” hop

$$\tau_2^- = \tau_2 \exp \left\{ \frac{\varepsilon_1 - \varepsilon_2}{T} \right\}. \quad (33)$$

We also note that in the regime $V \ll T$ the forward two-electron transition, illustrated with forward arrows in Fig. 2, acquires an activation energy, $\varepsilon_1 - \varepsilon_2 - V$, whereas the reverse two-electron transition $(1, r) \rightarrow (l, 2)$ becomes *activationless* with the characteristic time, τ_c , calculated as above. Therefore, the time of the forward two-electron transition is

$$\tau_c^+ = \tau_c \exp \left\{ \frac{\varepsilon_1 - \varepsilon_2 - V}{T} \right\}, \quad (34)$$

where τ_c is now given by

$$\frac{1}{\tau_c} = \frac{\Gamma_l \Gamma_r}{\pi \hbar U} \left[\Phi(\varepsilon_2/U) - \Phi(\varepsilon_1/U) \right], \quad (35)$$

with $\Phi(x)$ defined by Eq. (6). The latter expression was derived assuming that $(\varepsilon_1 - \varepsilon_2) \gg T$. As we will see below, in the ohmic regime, the most interesting situation corresponds to $(\varepsilon_1 - \varepsilon_2) \sim T \ll U$. Then one has to perform the integration in the general expression Eq. (4) [with ε_1 and ε_2 interchanged due to the redefinition of τ_c by Eq. (34)] using the explicit form of the distribution functions in the leads. This yields

$$\frac{1}{\tau_c} = \frac{\Gamma_l \Gamma_r U^2}{2\pi \hbar \varepsilon_1^2 (\varepsilon_1 + U)^2} \cdot \frac{\varepsilon_1 - \varepsilon_2}{1 - \exp[(\varepsilon_2 - \varepsilon_1)/T]}. \quad (36)$$

As in the previous section, we assume here that the sites are in the ratchet configuration, i.e. their energies are within the interval $[0, U]$ below the Fermi level, so that interaction-elevated energies of both sites are above the Fermi level. In the regime dominated by two-electron transitions, the only sets of the occupation numbers involved in the passage of current are $(0, 1)$ and $(1, 0)$. In the presence of reverse transitions, the master equation Eq. (20) assumes the form

$$\frac{dP_{1,0}}{dt} = -P_{1,0} \left(\frac{1}{\tau_c} + \frac{1}{\tau_2} \right) + P_{0,1} \left(\frac{1}{\tau_c^+} + \frac{1}{\tau_2^-} \right). \quad (37)$$

With the normalization condition, $P_{0,1} + P_{1,0} = 1$, this yields the following stationary solution for the probability $P_{0,1}$,

$$P_{0,1} = \frac{\tau_c^{-1} + \tau_2^{-1}}{\tau_c^{-1} + \tau_2^{-1} + (\tau_c^+)^{-1} + (\tau_2^-)^{-1}}. \quad (38)$$

In the presence of reverse hops, the stationary current can be calculated, *e.g.*, as the difference of the forward and reverse currents between sites 1 and 2, namely,

$I_c = P_{1,0}/\tau_2 - P_{0,1}/\tau_2^-$. With the use of Eq. (38), we then obtain

$$I_c = \frac{(\tau_c^+)^{-1} - \tau_c^{-1} (\tau_2/\tau_2^-)}{1 + (\tau_2/\tau_c) + (\tau_2/\tau_c^+) + (\tau_2/\tau_2^-)}. \quad (39)$$

In the ohmic regime, $V \ll T$, the numerator of Eq. (39) is proportional to V . Substituting (34) and (33) into (39), we find the ohmic resistance caused by two-electron hops

$$R_c^{-1}(T) = \frac{I_c}{V} = \frac{e}{T(\tau_2 + \tau_c)} \left[\exp \left(\frac{|\varepsilon_1 - \varepsilon_2|}{T} \right) + 1 \right]^{-1}. \quad (40)$$

Although in our derivation we assumed that $\varepsilon_1 > \varepsilon_2$, by introducing $|...|$ into Eq. (40) and also into Eq. (36), we ensure that it applies to $\varepsilon_1 < \varepsilon_2$ as well. Comparing this result to “non-ohmic” Eq. (21), we conclude that basically in the ohmic regime the current, I_c , acquires the activation energy, $|\varepsilon_1 - \varepsilon_2|$. We would like to emphasize that the necessary condition for both Eq. (21) and Eq. (40) to apply is that sites 1 and 2 are in the ratchet configuration.

While in the non-ohmic regime with $l \rightarrow 1$ and $2 \rightarrow r$ blocked, the two-electron hops constituted the *only* channel for activationless current flow, it is not obvious whether the “two-electron resistance” Eq. (40) can determine the net resistance in the ohmic regime. We address this question below.

Note first that, whether the current is governed by sequential hopping or by two-electron hops, the transfer of electrons between the leads necessarily includes the transition $1 \rightarrow 2$ (or $2 \rightarrow 1$). Thus, the “competition” between two-electron and sequential hopping is decided by the dominant mechanism of reoccupation of sites 1 and 2 *from the leads* during intervals *between* the hops $1 \rightarrow 2$ (or $2 \rightarrow 1$). If this dominant mechanism is two-electron transitions $(l, 2) \rightarrow (1, r)$ or reverse, then the longest waiting time for this transition, τ_c^+ , must be shorter than *all* characteristic single-electron times involved in the reoccupation of the sites. For single-electron transitions $l \leftrightarrow 1$ the shortest such time is given by

$$\ln \tau_1^\pm = \frac{2}{a} \left(\frac{D}{2} - d_1 \right) + \frac{\Delta_1}{T}, \quad (41)$$

where $\Delta_1 = \min \{ |\varepsilon_1|, |\varepsilon_1 + U| \}$ is the smallest activation energy of the hops $1 \rightarrow l$ (when site 2 is empty) and $l \rightarrow 1$ (when site 2 is occupied). Analogously, for transitions $r \leftrightarrow 2$, the shortest reoccupation time is defined as

$$\ln \tau_3^\pm = \frac{2}{a} \left(\frac{D}{2} - d_2 \right) + \frac{\Delta_2}{T}, \quad (42)$$

with $\Delta_2 = \min \{ |\varepsilon_2|, |\varepsilon_2 + U| \}$. The times (41) and (42) should be compared to the longest time of the two-electron transition, which in terms of d_1 and d_2 can be expressed as follows

$$\ln \tau_c^+ = \frac{2}{a} \left(D - d_1 - d_2 \right) + \frac{|\varepsilon_1 - \varepsilon_2|}{T}. \quad (43)$$

Then the conditions $\tau_c^+ < \tau_1^\pm$ and $\tau_c^+ < \tau_3^\pm$ can be cast in the form

$$\begin{aligned} d_2 &> \frac{D}{2} - \frac{a(\Delta_1 - |\varepsilon_1 - \varepsilon_2|)}{2T}, \\ d_1 &> \frac{D}{2} - \frac{a(\Delta_2 - |\varepsilon_1 - \varepsilon_2|)}{2T}. \end{aligned} \quad (44)$$

It is seen from Eq. (44) that the most favorable situation for two-electron transport to dominate is when the energies of the sites are close to each other, i.e. when $|\varepsilon_1 - \varepsilon_2| \ll |\varepsilon_1|, |\varepsilon_2|$. In this situation we have $\Delta_1 \approx \Delta_2 = \Delta$, so that the conditions (44) can be presented as $d_1, d_2 > [D/2 - a\Delta/2T]$. Graphically these conditions are represented by the vertical and horizontal lines on the plane $\{d_1, d_2\}$, see Fig. 6.

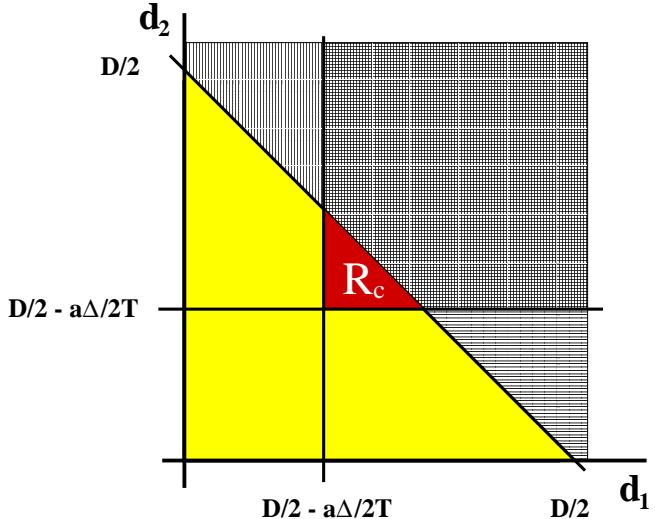


FIG. 6. (Color online) Triangular region defined by conditions $d_1, d_2 > D/2 - a\Delta/2T$ and $d_1 + d_2 < D/2$ indicates the site positions where two-electron hops dominate the transport.

In the domain where these conditions are met, Eq. (40) applies. Note, however, that when τ_2 is much longer than τ_c , it is the hop $1 \longleftrightarrow 2$ that determines the resistance regardless of whether the reoccupation of sites 1 and 2 occurs via two-electron or single-electron transitions. Thus, the “true” two-electron-hopping regime is realized when $\tau_2 < \tau_c$. The latter condition can be presented as $d_1 + d_2 < D/2$. It corresponds to the diagonal line in Fig. 6. This line together with the horizontal and vertical lines define a non-empty domain of site positions d_1 and d_2 if the condition

$$D < \frac{2a\Delta}{T} \quad (45)$$

is met. The condition (45) has a transparent physical meaning. Single-electron hops require shorter tunneling distances, but involve activation, whereas two-electron hops require tunneling over longer distances, but they are practically activationless when $\varepsilon_1 \approx \varepsilon_2$. Thus, in accordance with Eq. (45), the lower the temperature, the wider is the range of the barrier thicknesses where two-electron hops dominate the transport.

According to the definition of Δ as the minimal of the activation energies of the hop onto (or out of) Coulomb-shifted and unshifted sites, its value cannot exceed $U/2$. Then the most favorable situation for two-electron-dominated hops is $\varepsilon_1 \approx \varepsilon_2 \approx -U/2$. In this situation, Eq. (45) can be presented as $D < aU/T$.

V. CONCLUDING REMARKS

In the present paper we studied the effect of interactions on inelastic transport through a two-site chain. In particular, we have identified the domain of energies and positions of sites where the transport occurs via two-electron transitions, which are impossible without interactions. In the ohmic regime, the most favorable situation for the two-electron transport regime realizes when the energies of the sites are close, $\varepsilon_1 \approx \varepsilon_2$. Note, however, that throughout the paper we assumed that the difference between the site energies, as well as their energy distance to the Fermi level, exceed the site widths, Γ_l, Γ_r . This ensures that there is no resonant tunneling of a single electron via the two-site configuration². While *interaction-induced* resonant tunneling with Γ_l, Γ_r determined by *two-electron* virtual transitions²⁶ does not require complete alignment of site energies, it does require more than two sites.

For a thick barrier with localized states the current is governed by impurity chains containing many sites^{27,28}. Their energies are close to the Fermi level, and they are almost equidistant. We do not expect interactions to have a strong effect on hopping through these “optimal” chains for the following reason. While the waiting times for all hops constituting the chain are approximately the same, the net resistance is determined by the longest of these times. This is reminiscent of the two-site chain, considered above, with τ_2 longer than τ_1, τ_3 , and τ_c . As we saw before, under this condition, the fashion (correlated or non-correlated) in which electrons were delivered to/from the sites of the main hop was not important.

Unlike inelastic cotunneling, the regime of *elastic* cotunneling through a quantum dot²⁴ does not have an analog within the two-site model. This is because in the case of a quantum dot an electron can be transferred between the left and right leads only via the dot. By contrast, in the two-site model, an electron can tunnel directly between the leads; the amplitude of this process exceeds the amplitude of elastic cotunneling via each of the two sites.

Note also, that the two-site model is too simplistic to capture another interaction effect on hopping transport, which is the effect of distant “fluctuators” away from the current-carrying path on the passage of current²⁹.

There exists certain analogy between the regime of transport by two-electron hops via impurities considered here and transport by cotunneling through granular arrays^{30–32}. The important difference is, however, that in Refs. 30–32 each granule is assumed to contain a large number of levels; cotunneling proceeds via these levels, and it is the finite charging energy of an *individual* granule that allows this process. Interaction *between the granules* is essentially irrelevant in calculating the rates of hops^{30–32}.

Since we consider *sites* rather than granules, it is the on-site Hubbard interaction, U_0 , that plays the role of charging energy. This interaction is assumed infinite in our consideration. On the other hand, the *intersite* interaction plays a central role for transport. As a consequence of the fact that two-electron transitions in a two-site model are due to the intersite interaction, the net rate of a hop is, essentially, the *sum* of the rates for two-electron and subsequent single-electron transitions. By contrast, in the case of granules, the probability of a particular hop between two distant granules is proportional to the *product* of tunneling probabilities between *all* adjacent granules along the path.

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